

Remarks

Applicants respectfully request the Examiner to reconsider the present application in view of the foregoing amendments to the claims and the following remarks.

The Office Action is non-final. Claims 1-4 and 6-13 are currently pending in the present application. Claims 1 and 9 have been amended to further clarify and define the invention. Claim 1 has been amended to incorporate the textual subject matter of claim 5, now cancelled. Claims 12 and 13 are new. Support for claim 12 is based on original claims 1 and 2. Support for claim 13 is based on original claims 1 and 8.

Entry of the present Amendment is respectfully requested.

Rejections under 35 U.S.C. § 103(a)

The following rejections under 35 U.S.C. § 103(a) are presented by the Examiner.

Claims 1-11 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Kwon *et al.*, WO 02/083557 (hereinafter "Kwon") in view of Sonobe *et al.*, U.S. Patent No. 5,587,255 (hereinafter "Sonobe '255"), in view of Sonobe *et al.*, U.S. Patent No. 5,616,436 (hereinafter "Sonobe '436").

Claim 3 stands rejected under 35 U.S.C. § 103(a) as being unpatentable over Kwon, Sonobe '255, Sonobe '436, as applied to claim 1, and further in view of Nagamine *et al.*, U.S. Patent No. 5,932,373 (hereinafter "Nagamine").

Claims 7, 10 and 11 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Kwon, Sonobe '255, Sonobe '436, as applied to claim 1, and further in view of Yoon *et al.*, U.S. Patent No. 6,218,050 (hereinafter "Yoon").

Claim 8 stands rejected under 35 U.S.C. § 103(a) as being unpatentable over Kwon, Sonobe '255, Sonobe '436, as applied to claim 1, and further in view of Sonobe *et al.*, U.S. Patent No. 5,527,643 (hereinafter "Sonobe '643") and Lu *et al.*, "Anodic Performance of Vapor-

Derived Carbon Filaments in Lithium-Ion Secondary Battery,” Carbon, Vol. 39, pp. 493-496 (2001) (hereinafter “Lu”).

Applicants respectfully traverse the above rejections.

Based on the following, Applicants contend that the Examiner’s position for each rejection is unsupportable, thereby making the presently claimed invention unobvious over the cited references in each of the rejections above.

The Present Invention

As recited in amended claim 1, the negative electrode material for non-aqueous electrolyte secondary batteries of the present invention, comprises: a spherical carbon material, first characterized by having a sphericity of at least 0.8. The electrode material is a non-graphitizable carbon material as represented by an average (002) interlayer spacing d_{002} of 0.365 - 0.400 nm, and a crystallite size in a c-axis direction $L_{c(002)}$ of 1.0 - 3.0 nm, as measured by X-ray diffractometry; has been sufficiently carbonized as represented by a hydrogen-to-carbon atomic ratio (H/C) of at most 0.1 as measured by elementary analysis; and has a small particle size as represented by an average particle size Dv_{50} of 1-20 μm .

The electrode material is further characterized by an appropriate level of product $S \times Dv_{50}$ of 3 – 40. This limitation is significant in providing a balance of a small specific surface area effective for preventing decomposition of the electrolytic solution in non-aqueous electrolyte secondary batteries, and doping-dedoping performances of the batteries. The significance of this limitation is discussed in more detail on page 10, line 22, to page 11, line 12, of the present specification and is herein reproduced:

“[0020] In order to suppress the decomposition of an electrolyte solution, it is preferred to provide a smaller specific surface area. However, the average particle size and the specific surface area are in a relationship of mutually inverse proportion, and a smaller average particle size tends to result in a larger specific surface area. In the present invention, by providing the negative electrode material with a shape of sphere which can minimize the specific surface area per volume and also by

optimization of the texture and controlling the surface structure of the synthetic resin as a carbon precursor, it has become possible to provide a remarkably smaller specific surface area even at a particle size smaller than that of a negative electrode material obtained through pulverization. From this viewpoint, it is preferred to provide as small a product of specific surface area S (m^2/g) and average particle size Dv_{50} as possible. On the other hand, too small a product of specific surface area S (m^2/g) and average particle size Dv_{50} leads to undesirable obstruction of the doping-dedoping reactions of lithium. The product of S (m^2/g) and Dv_{50} is preferably 3 - 40, further preferably 3 - 30."

Differences between the Invention and the Cited References

Kwon

Kwon discloses a method for producing spherical carbon usable as a negative electrode material for non-aqueous electrolyte secondary batteries. Kwon discloses a method of mixing a carbon precursor with an inorganic dispersion media (first method) or a silicone oil dispersion media (second method), followed by carbonization of the carbon precursor before removal of the inorganic dispersion media (first method) or after removal of the silicone oil dispersion media (second method) (See Kwon, page 11, lines 4-17). The Kwon reference discloses resin, pitch or a mixture thereof as a preferable carbon precursor (See page 11, line 21- page 12, line 1). As the resin, a thermosetting resin is preferred, inclusive of phenolic resins, furan resin, epoxy resin, polyacrylonitrile resin, polyimide resin, polyphenylene resin, biphenol resin, divinyl styrene copolymer, cellulose and mixture thereof (See page 12, lines 4-8), among which only phenolic resin is used in the working examples.

While phenolic resin, actually revealed as a useful carbon precursor, may be used as a starting material for providing a spherical carbon material of a non-graphitizable carbon as intended by the present invention, it provides a negative electrode material showing an excessively large specific surface area S of over $30 \text{ m}^2/\text{g}$ giving a product $S \times Dv_{50}$ of over 420. This is much larger than 3-40 in amended claim 1 of the present application, as shown in Table 2, page 39, of the present specification. Thus, this leads to a noticeable decomposition of

electrolytic solution as discussed above.

Kwon does not teach or suggest the significance of an appropriate level of product $S \times Dv_{50}$ of 3 - 40 for providing improved electrode performances.

Applicants note that spherical carbon particles formed by carbonization of a phenolic resin exhibit inferior electrode performance as shown in Comparative Examples 7 and 8 of the present specification, and as represented by a low (charge-discharge) efficiency of 76 % and a low cycle performance of below 70 % after a 100-cycle test (See paragraph [0094], pages 36-37, and Table 2, page 39, of the present specification).

Kwon *per se* discloses a remarkably inferior charge-discharge efficiency of about 70 % in a first cycle (See Kwon, Fig.6). Although cycle performance during the second to tenth cycle appears to be stable (Fig.6), no severe cycle performance tests were performed as in the present invention, which performed a 100-cycle test.

Sonobe '255 and Sonobe '436

Sonobe '255 and Sonobe '436 disclose negative electrode materials of non-graphitizable carbon, but fail to disclose electrode materials comprising spherical carbon particles having a particle size Dv_{50} of 1-20 μm suitable as an electrode material. Thus, the problem of the pulverized electrode materials of the prior art as discussed on page 3, paragraph [0007], of the present specification, has not been solved.

More specifically, Sonobe '255 produced a spherical carbon material having an average particle size of 400 μm and a specific surface area S of 2.4 m^2/g in Example 1 (See Sonobe '255, column 11, lines 7 - 13), but the spherical carbon material was pulverized into an average particle size Dv_{50} of about 20 μm for use as an electrode material (See column 13, lines 62 - 66) giving a product $S \times Dv_{50}$ of 48.

In Sonobe '436, pulverized carbon precursor particles of approximately 25 μm

(Examples 1-3) or 20 μm (Examples 4 and 5) were further carbonized to provide carbon electrode materials (See Sonobe '436, column 9, line 62, to column 11, line 3)

As is understood from the above discussion, both Sonobe '255 and Sonobe '436 also fail to disclose the significance of an appropriate level of product $S \times D_{v50}$ of 3-40 for providing improved electrode performance, thus failing to remedy the deficiencies of the Kwon reference with regard to amended claim 1.

Further Distinguishing Features of the Present Invention

Additionally, Applicants point out that new claim 12 recites another characteristic feature of the spherical carbon electrode material of the presently claimed invention, namely, that it is a carbonization product of a vinyl resin.

As already discussed above, Kwon discloses a thermosetting resin as a preferred carbon precursor, inclusive of phenolic resins, furan resin, epoxy resin, polyacrylonitrile resin, polyimide resin, polyphenylene resin, biphenol resin, divinyl styrene copolymer, cellulose and mixture thereof (See Kwon, page 12, lines 4-8), among which only phenolic resin is used in the working examples.

In the above, polyacrylonitrile resin and divinyl styrene copolymer are included as thermosetting resins. Applicants submit that this is incorrect. Polyacrylonitrile resin is a thermo-plastic vinyl resin, and divinyl styrene copolymer is a vinyl resin (as acknowledged by the Examiner) which can be thermoplastic at a low divinyl-benzene content or non-thermoplastic at a high divinylbenzene content. These vinyl resins cannot be made spherical similarly as a phenolic resin according to the above-discussed first or second method of Kwon, due to fusion or decomposition in the carbonization step. Thus, Kwon substantially fails to disclose spherical carbonization product of a vinyl resin.

In contrast, in the presently claimed invention, the spherical carbonization product of a

vinyl resin is preferably formed through a suspension polymerization in an aqueous dispersion medium to form a spherical vinyl resin (See paragraph [0026]), followed by infusibilization at 150 – 400 °C in an oxidizing atmosphere and carbonization in a non-oxidizing atmosphere (See paragraph [0024]). The infusibilization is required since a vinyl resin, unlike a thermosetting resin, is simply melted or decomposed to lose its spherical shape if directly subjected to carbonization.

Sonobe '255 and Sonobe '436 disclose negative electrode materials of non-graphitizable carbon of pitch origin, but fail to disclose spherical carbon electrode materials (as discussed above), particularly those formed by carbonization of a vinyl resin, thus failing to remedy the deficiencies of Kwon in suggesting new claim 12.

With regard to new claim 13, it recites another characteristic feature of the spherical carbon electrode material of the present invention, which is that it has a nitrogen content of 0.5 - 5wt. %.

As discussed in paragraph [0021] (See page 11, lines 13-16) of the present specification, it is effective to incorporate such a small amount of nitrogen in the spherical carbon electrode material of the present invention in order to provide a relatively small specific surface area, thus providing an appropriate level of product $S \times Dv_{50}$. This is clear from Example 7, which is lacking nitrogen (N) content and provides a large specific surface area of over 30 m²/g and a large product $S \times Dv_{50}$ of over 240 (See Table 2, page 39, of the present specification).

Therefore the Kwon, Sonobe '255 and Sonobe '436 references all fail to teach or suggest the inclusion of such a small amount of nitrogen in spherical carbon electrode materials.

In light of the above arguments, Applicants submit that the assertions made by the Examiner regarding the Kwon, Sonobe '255 and Sonobe '436 references lack merit, thus making the Examiner's position unsustainable.

Accordingly, since all of the above rejections are based on the Kwon, Sonobe '255 and Sonobe '436 references, the present invention is not obvious to one of ordinary skill in the art. As discussed, the secondary references, Sonobe '255 and Sonobe '436, fail to remedy the deficiencies of Kwon.

Regarding the additional secondary references, Nagamine, Yoon, Sonobe '643 and Lu, these references fail to remedy the deficiencies of Kwon, Sonobe '255 and Sonobe '436, outlined above. Therefore, even if the references were combined in the manner asserted by the Examiner, the result of such combination would still not suggest the claimed invention.

Applicants respectfully request reconsideration and withdrawal of the above rejections.

Conclusion


Applicants respectfully submit that all of the rejections raised by the Examiner have been overcome, and that the present application now stands in condition for allowance.

Should there be any outstanding matters that need to be resolved, the Examiner is respectfully requested to contact Paul D. Pyla at the telephone number below, in an effort to expedite prosecution in connection with the present application.

If necessary, the Commissioner is hereby authorized to charge payment or credit any overpayment to Deposit Account No. 23-0975 for any additional fees required under 37 C.F.R. §§1.16 or 1.17.

Respectfully submitted,

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